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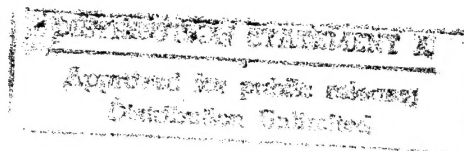
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UNITED STATES ATOMIC ENERGY COMMISSION

STUDIES OF THE ALUMINUM-URANIUM
ALLOYING REACTION

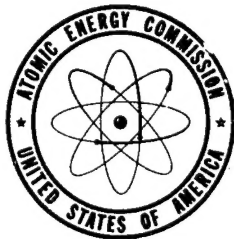
By
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December 15, 1949

Brookhaven National Laboratory
Upton, New York

Technical Information Service, Oak Ridge, Tennessee



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D.W.Bareis

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ABSTRACT

It was found in this investigation that an alloying reaction occurred wherever and whenever clean metallic surfaces of aluminum and uranium were brought into contact within the temperature range of 250°C to 450°C. Anodization of the aluminum surface prevented the alloying reaction. The effects of temperature, aluminum purity, and pressure between the metal surfaces on the alloying reaction were studied qualitatively. The alloying reaction produced UAl_3 , which appeared to be formed by the diffusion of the uranium through the UAl_3 layer.

INTRODUCTION

Early work at the Chicago Metallurgy Laboratory indicated that, when aluminum and uranium were brought together under certain conditions, a diffusion type reaction between the two surfaces produced alloys of aluminum and uranium accompanied by an increase in volume. When the Clinton reactor was designed, it was thought that this reaction would not limit the life of the fuel elements which were to have a maximum interface temperature of 250°C. An investigation into the causes of the recent Clinton slug ruptures has shown that as many as one third might have been caused by the penetration of the aluminum jacket through an alloy formation.¹ The Brookhaven reactor fuel elements have an anodized surface on the aluminum in contact with the uranium. Recent tests have shown that an anodized coating on the aluminum will prevent the alloy formation at temperatures well above the maximum design value of 350°C for the metal interface of the Brookhaven reactor fuel elements.²

The present work was undertaken to determine what conditions are necessary to promote the alloy formation, and to study the mechanism of the reaction. Experience gained when studying the effects of iodine vapor on a canned slug indicated that reproducible results could not be attained by using canned slugs.³ It was decided to use a sandwich type arrangement of aluminum and uranium disks, held together by a stainless steel clamp. The sandwich assembly was sealed within a pyrex container to allow for the continuous observation of the assembly and to contain the atmosphere surrounding the assembly. The conditions which were varied in this investigation are listed below:

1. The temperature of the test furnace.
2. The purity of the aluminum disks.
3. The treatment of the aluminum disk surface.
4. The distance between the aluminum and the uranium disk surfaces.
5. The atmosphere surrounding the assembly.
6. The pressure on the aluminum and the uranium disks.
7. The residence time in the furnace.

Since it was not known at the start of this investigation when the alloying reaction would occur and whether a reaction would occur, it was not known what the reaction rate would be. The series of tests reported herein was made to obtain primarily qualitative results. Fortunately, the first assemblies tested underwent extensive alloying reactions, and thereby indicated the magnitude of, and the temperature effect on, the reaction rate. The results of this investigation indicated which conditions must be met to obtain the alloying reaction. More quantitative results are needed to obtain a better understanding of the reaction mechanism.

EXPERIMENTAL PROCEDURES

Preparation of Assemblies

A brief description of the apparatus and the procedure used is given here; the details are presented in the Appendix.

The apparatus and procedure used in this investigation provided the following conditions:

1. Produced clean, smooth, and parallel surfaces on the aluminum and uranium disks.
2. Brought the aluminum and uranium surfaces together without contamination or reaction with air.
3. Kept surfaces together until completion of the test.
4. Maintained a vacuum or controlled atmosphere around the assembly.
5. Regulated the distance between the aluminum and uranium surfaces.
6. Enabled the observation of the assembly continuously without disturbing the test conditions.

Figure 1 shows a typical sandwich assembly under test conditions. Figure 2 shows an exploded view of the various components used in the assemblies. Not all of these components were used in every assembly. The components are listed and described in Table I.

The uranium and aluminum surfaces were machined parallel and smooth to a tolerance of ± 0.1 mil. The disks were then polished metallographically and stored in absolute alcohol. The various components were cleaned and stored in alcohol. The components were dried and clamped together under a purified helium atmosphere. Then they were placed within the pyrex tube, and the tube was sealed at the point marked "A" in Figure 2, under a helium atmosphere. The pyrex container was evacuated for 15 hours, and then sealed at the point marked "B" in Figure 2.

Test Procedure

The completed assembly was placed in a semicircular trough lined with asbestos paper. A thermocouple, attached to a recorder, was placed under the assembly. The unit was brought up to the desired temperature in a preheat furnace and then placed in a constant temperature tube furnace for the test. The assembly was observed periodically by photographing the unit as it was withdrawn momentarily from the furnace. This process required removing the unit for a period of less than 5 seconds, which had no effect on the test conditions, but allowed a permanent record to be made of the progress of the test. After a definite reaction had been detected, the assembly was removed from the furnace. The reaction usually forced the aluminum and uranium disks apart, as was readily shown by the photographs. In the cases where no detectable reaction occurred, the assembly was removed after sufficient time had elapsed for the reaction to have occurred.

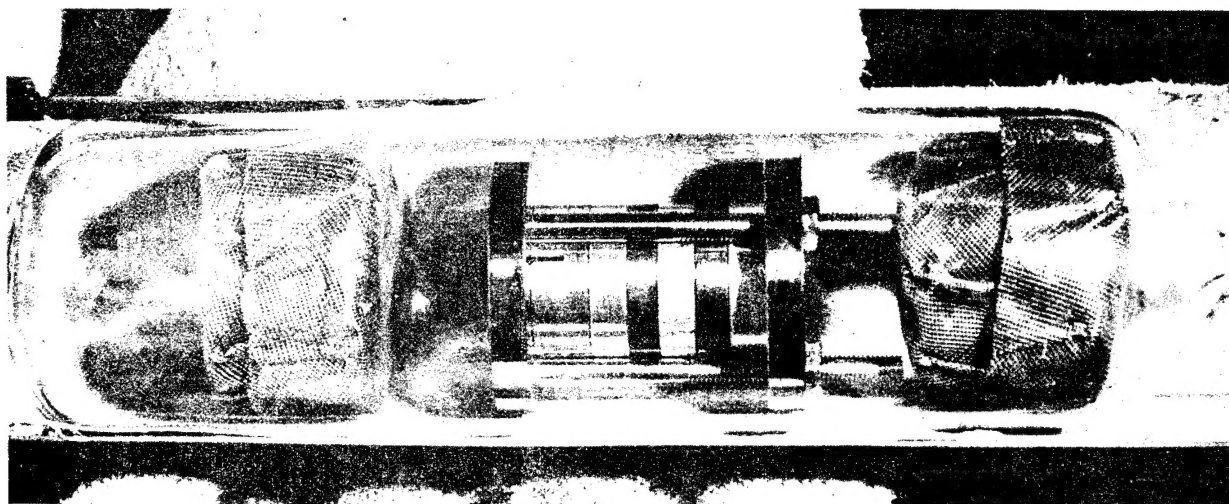


Figure 1. A sandwich assembly (#3).

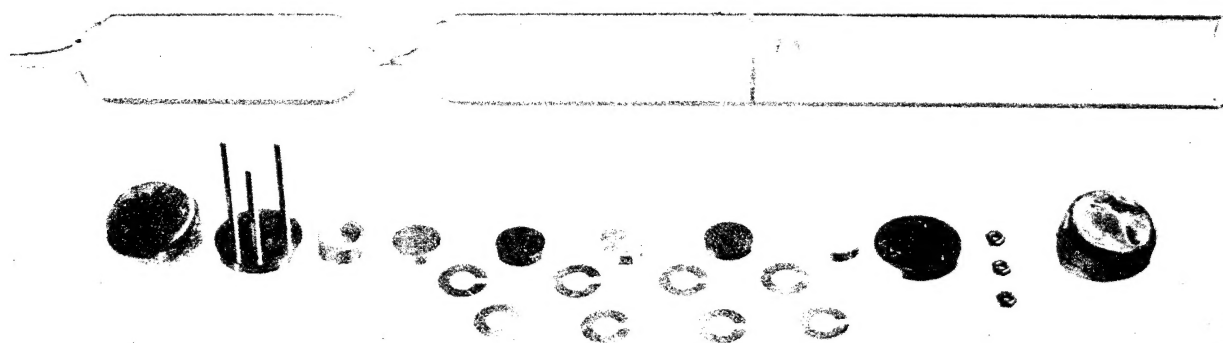


Figure 2. The assembly components.

At the completion of the test, the pyrex container was broken, and the sandwich was disassembled. The condition of the disks was recorded, and photographs of the surfaces were made.

Table I			
Sandwich Assembly Components			
Component	Material	Size	Use of Component
Container	Pyrex	50 mm ID	Contained assembly and surrounding atmosphere.
Scavenger Baskets	Nickel Screen and Uranium Turnings		Removed O ₂ and N ₂ from atmosphere around assembly.
Clamp	Stainless Steel		Held disks together.
Disks	Aluminum and Uranium	1" diameter 1/4" thick	Material under test.
Shims	Stainless Steel	1" OD, 1/2" ID, 1 mil thick	Separated aluminum and uranium surfaces.
Compression Cup	Aluminum	1" OD, 3/8" thick; hole: 3/4" OD x 1/4" deep	Relieved excessive pressures from thermal expansion and alloy growth.

Future Work

It is tentatively planned to obtain aluminum penetration rate data by a different procedure. Uranium foil will be cleaned and pressure sealed between polished aluminum disks in an inert atmosphere. This will enable the handling of the assembly in the air. It will also allow for a rapid heating and cooling period for the very short high temperature exposures. A thermocouple will be imbedded in the aluminum disk and connected to a rapidly recording potentiometer. The assembly will be cut normal to the uranium surface for the examination of the alloying reaction.

Table II

Conditions of the Sandwich Assemblies

Number	Type Aluminum	Clearance (mils)	Seal Off Pressure (microns Hg)	Atmosphere	Furnace Temp. (°C)	Furnace Residence Time (hours)	Results
1	2S	0, 1, 2	22.0	Vacuum	400	151	Extensive alloying reaction.
2	2S	0, 1, 2	1.5	"	450	31	Extensive alloying reaction.
3	99.99%	0	5.0	"	400	148	Moderate alloying reaction.
4	"	0	7.0	"	350	455	Moderate alloying reaction.
5	"	0	6.0	"	450	27	Extensive alloying reaction.
6	"	0	5.0	"	250	1800	Slight alloying reaction.
7	"	0	2.0	"	450	28	Limited area alloying reaction. [Al on U, and U on Al; no clamp used.]
8	"	5	3.0	"	450	99	No alloying reaction.
9	"	5	-	Air	450	198	No alloying reaction (extensive oxidation).
10	"	5	-	Helium	425	197	No alloying reaction (slight corrosion).
11	"	0	1.5	Vacuum	400	149	Moderate alloying reaction.
12	"	0	20.0	"	450	792	No alloying reaction (Al surface anodized).

RESULTS

The sandwich assembly conditions which were varied in this investigation are listed in the Introduction. The conditions used for the individual sandwich assemblies are given in Table II.

Description of Alloying Reaction

The reaction can be portrayed very effectively by means of photographs of the metal surfaces. Figures 3 through 6 show the various stages of the reaction.

The preliminary stage (Figure 4) left the disks held very tightly together. The grain structure was usually apparent when the disks were parted. The surface of the aluminum was depressed over the area of the reaction, while the uranium surface was raised correspondingly; this could only be determined by the use of a microscope, since the surfaces actually felt and looked smooth.

The intermediate stage (Figure 5) was accompanied by the initial formation of the often referred to "blisters," i.e., the alloy formed in the reaction was raised from the uranium surface, so that a void space existed between the uranium and the alloy. The "blisters" were cone-shaped, with the apex embedded in the aluminum. The alloy had a dull gray color. The disks were parted with some difficulty. The alloy was very brittle; it broke near the uranium surface, being held mechanically by the aluminum. On the other hand, the alloy appeared to be held very tenaciously to the uranium surface. When the alloy was parted from the aluminum surface, a thin coat of a black material was visible.

The advanced stage (Figure 6) of alloy formation, or "blistering," forced the aluminum and uranium surfaces apart as much as 1/16 inch. The aluminum compression cup and the aluminum disks were visibly deformed. It was very apparent that the alloy was much less ductile than the aluminum. The uranium and the aluminum disks were separated easily, with the alloy again breaking off near the uranium surface. The alloy cones, or "blisters," which were formed were very thin, consisting mostly of void space. The alloy expansion was almost entirely into the aluminum, there being only a slight depression in the uranium surface below the "blisters." The calculated increase in volume due to the UAl_3 alloy formation, without the void space, was only 17%. This increase in volume of the alloy could only be attained by movement into the softer aluminum surface. If one assumes that the alloy forms a cone, the base angle would have to be approximately 20° , which indicates that the blister formation could have been caused by the volume expansion of the alloy formed in the reaction.

A better picture of the penetration into an aluminum disk is shown in Figure 7. The reaction occurred at the center of this disk, since the stainless steel shims which were used to separate the surfaces failed to prevent a contact at the center.

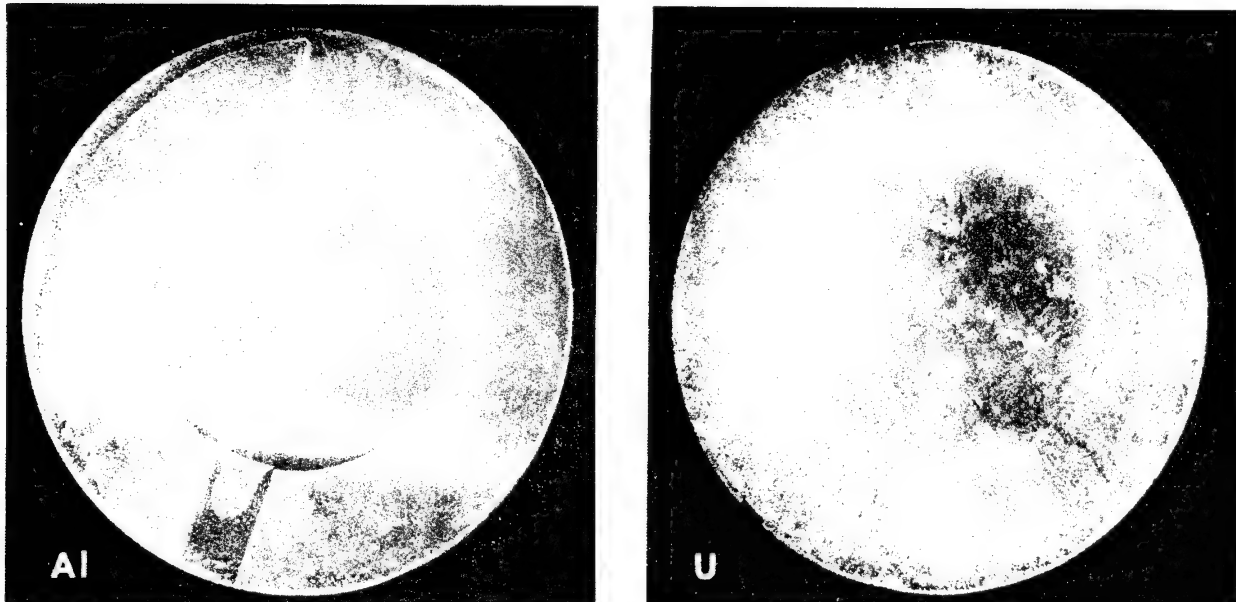


Figure 3. Al and U surfaces after the test. No reaction - surfaces separated (Interface #10, Assembly #1).

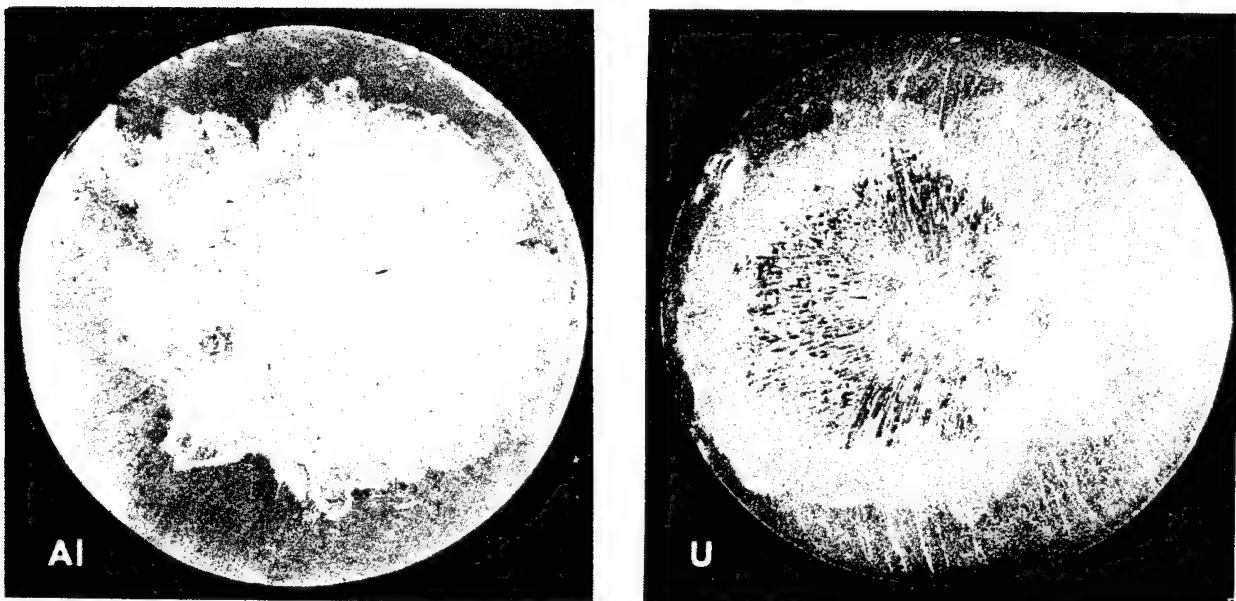


Figure 4. Al and U surfaces after the test. Grain structure visible within contact area (Interface #2, Assembly #4).



Figure 5. Al and U surfaces after the test. Grain structure plus isolated "blisters" (Interface #2, Assembly #5).

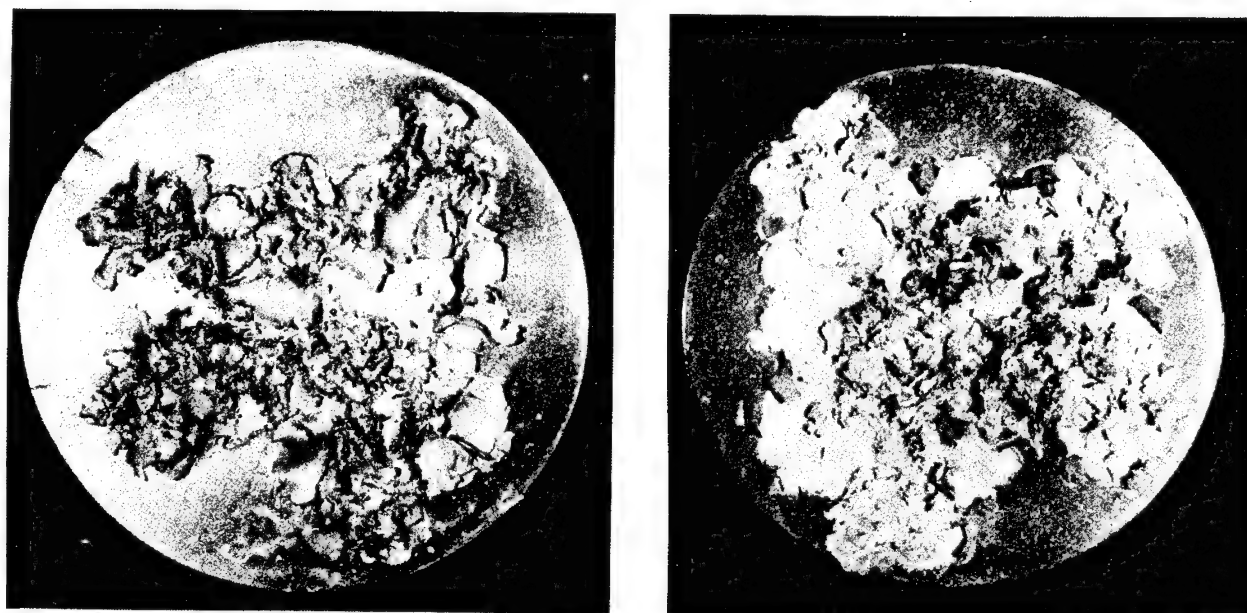
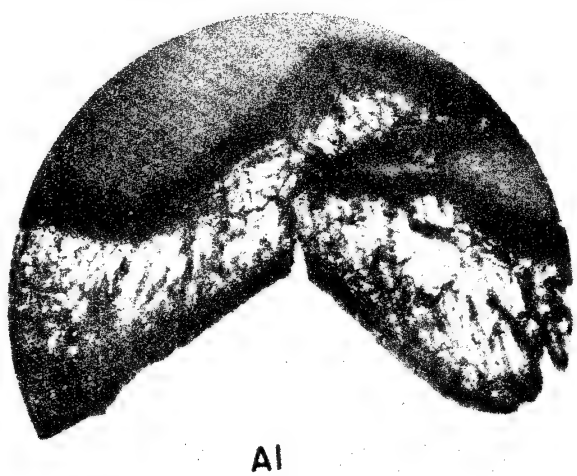


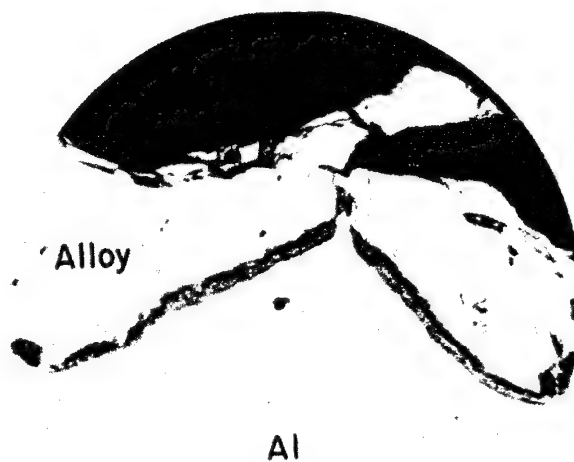
Figure 6. Al and U surfaces after the test. "Blister" formation over contact area (Interface #2, Assembly #1).



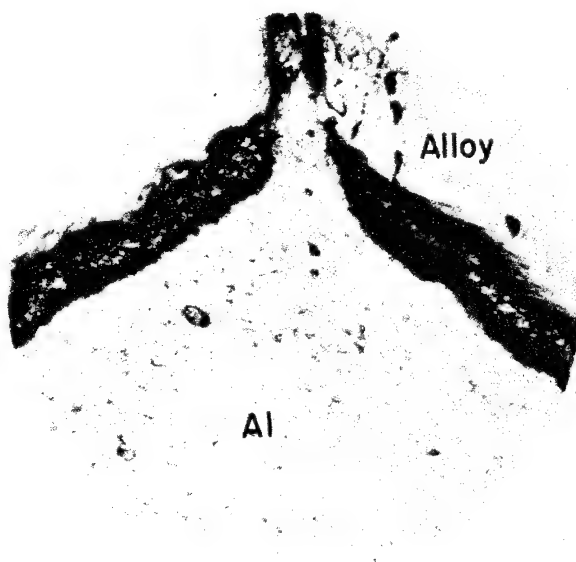
Figure 7. Cross section of an aluminum disk
(8X, HF etch) (Assembly #1).



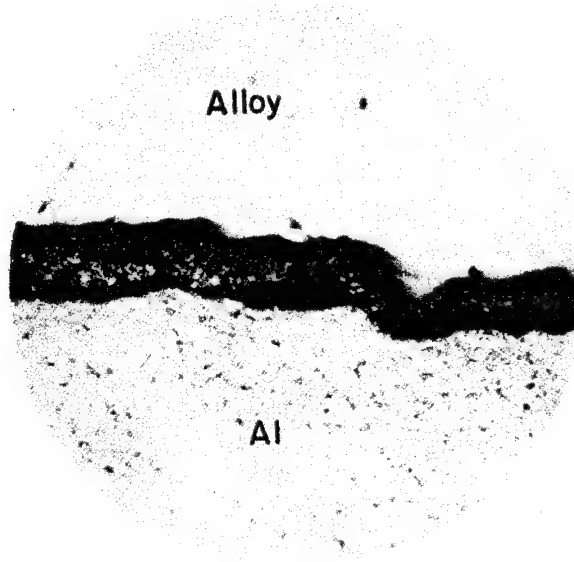
(a) Alloy on aluminum surface
(150X, HF etch).



(b) Alloy on aluminum surface
(150X, no etch).



(c) Black substance between alloy and alu-
minum (500X, no etch).



(d) Black substance between alloy and alu-
minum (500X, no etch).

Figure 8. Enlarged cross section of an aluminum disk (Assembly #1).

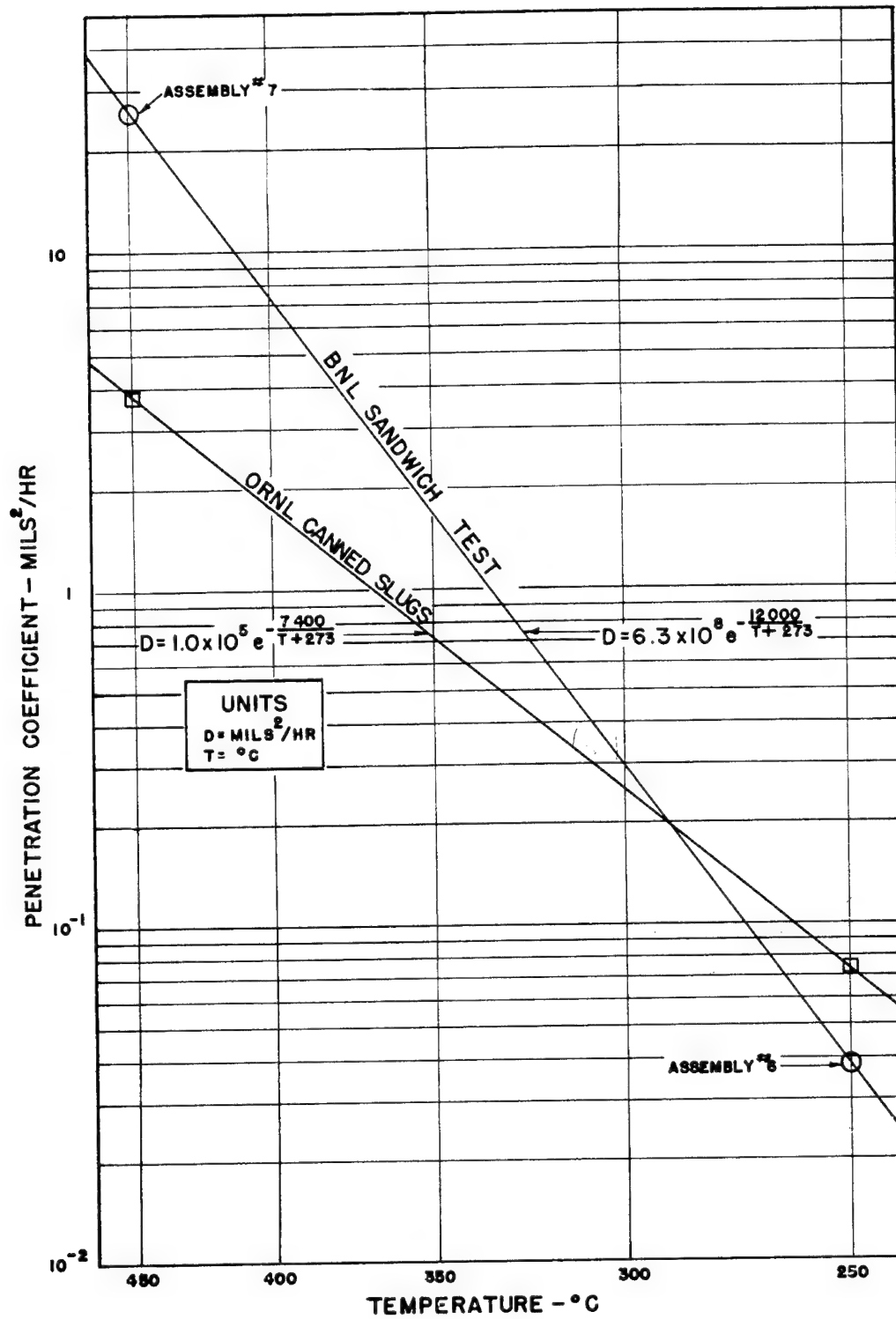


Figure 9. Variation of the maximum aluminum penetration rate with the temperature using BNL and ORNL data.

At the point marked "A" is a smooth round which was formed when the shim was forced into the aluminum by the alloying reaction. The darker material at the aluminum surface is the alloy. Figure 8 gives a greater enlargement to portions of the alloy-aluminum interface. The black material located at the alloy-aluminum interface is very evident in all of these pictures. This substance had a very uniform thickness, which was in the order of 1 mil.

Effects of Varying Test Conditions

1. Temperature

The temperature range of this investigation was 250°C to 450°C. Most of the assemblies were tested at the high temperatures, where faster reaction rates were obtained, thus decreasing the time required for the test. Qualitatively, the alloying reaction rate increased with increasing temperatures. In order to obtain reaction rate data from the test results, it was necessary to eliminate all the assemblies except #6 and #7 from consideration, because there was either no reaction, or the reaction had reached a very advanced stage. The latter caused deformation in the aluminum, and thus made the measurement of the depth of penetration into the aluminum unreliable. Assembly #6 had a very slight reaction, the rate being low at 250°C. Assembly #7 with the uranium disk resting on top of the aluminum had no deformation of the aluminum and only one reaction point of very moderate depth. In the latter case, the temperature was 450°C. The point of maximum aluminum penetration was measured by observing the aluminum disk microscopically perpendicular to the direction of the diffusion. Observations were made intermittently with the polishing of the mounted section until the maximum depth was passed or removed. Assemblies #6 and #7 had maximum aluminum penetrations of 8.27 mils and 26.7 mils respectively. The measurements were plotted in Figure 9 as the aluminum penetration depth squared divided by the time for the test versus the reciprocal of the absolute temperature of the test. This is the usual method of plotting diffusion data in order to obtain a straight line relation. As yet, there is insufficient data to determine whether or not a linear relationship exists for the alloying reaction, which could conceivably depend on several diffusion rates and/or reaction rates over a moderate temperature range. For a comparison, values given in an Oak Ridge report¹ are also plotted in Figure 9. These latter values were obtained from studies of canned slugs, and represent the maximum aluminum penetration rate found at 250°C and 450°C. Using a can thickness of 30 mils, it was possible to calculate the minimum time required for the penetration of the aluminum by the alloying reaction from the above data. These calculated values are shown in Table III.

2. Aluminum Purity

Two grades of aluminum were used for this investigation. Assemblies #1 and #2 contained disks made from commercial 2S aluminum bar stock. The remaining assemblies contained 99.99% pure aluminum. The pure aluminum was obtained from Aluminum Company of America in the ingot form, and was then vacuum cast at Massachusetts Institute of Technology into a form suitable for machining. Spectrographically, there was no difference between the pure aluminum in the ingot form

and in the vacuum castings. The actual analyses are given in the Appendix. It appeared from an examination of the alloy formation that the 2S aluminum had a slightly higher reaction rate. No quantitative data are available.

Table III		
Minimum Time Required to Penetrate 30 Mils of Aluminum by the Alloying Reaction		
Temperature	BNL Sandwich Assemblies	ORNL Canned Slugs
250°C	990 days	510 days
300°C	130 days	150 days
350°C	22 days	52 days
400°C	130 hours	21 days
450°C	35 hours	10 days

3. Aluminum Surface Treatment

All the assemblies contained aluminum disks with no oxide on the surface, except Assembly #12, which contained anodized aluminum disks. This last assembly was tested for a period which was 30 times as long as that required to give an extensive reaction with no anodization. The temperature of the test was 450°C. No reaction was detected in this assembly. Anodization of the aluminum surface appears to prevent the alloying reaction.

4. Surface Contact

It was found that a contact between the aluminum and the uranium was necessary before the alloying reaction could proceed. In Assemblies #1 and #2, an interesting incident was noted. The reaction expansion caused some of the separated surfaces to come into contact and react. No reaction was found in the other assemblies containing separated surfaces.

5. Atmosphere

All the assemblies were sealed in a vacuum except Assemblies #9 and #10 which contained air and helium respectively. These assemblies also had separated surfaces. Assembly #9 showed extensive oxidation of the uranium and stainless steel spacers, while Assembly #10 showed only a very slight corrosion of the uranium. The combined effects of the atmosphere and surface separation produced no alloying reaction in these assemblies. No test was made with a blanket atmosphere in an assembly having surface contact.

6. Pressure

No quantitative effect of pressure on the reaction rate was obtained, since the pressure on the disks in an assembly was indeterminable. The pressure between the aluminum and the uranium was dependent on the initial pressure, the type of aluminum present, the temperature of the test, and the rate and the nature of the alloy formation. Assembly #7 contained a disk of aluminum resting on the uranium and a disk of uranium resting on the aluminum. In both cases the alloying reaction occurred, but not to the same extent as in the clamped assemblies. The area of the reaction was small in the case of the former because the initial contact area required to support the weight of the disks was small.

7. Time

The residence time of the assemblies in the furnaces was dependent on the reaction rate; or, in those cases where no reaction occurred, the period was lengthened to several times that used for the assemblies showing a positive reaction at the corresponding temperatures. As mentioned previously, the first assemblies tested had a positive reaction and indicated the relative effect of temperature and furnace residence time.

Mechanism of the Alloying Reaction

When the assemblies with large amounts of alloy formation were taken apart, some of the alloy broke off the disks. It can be assumed that most of this alloy came from near the aluminum surface, since the alloy was less firmly attached to the aluminum. Also, some of the alloy attached to the uranium disks was pried off and collected. Portions from both of the collected samples were analyzed by the Chemistry Department for aluminum and uranium. Corresponding portions were used to obtain X-ray diffraction patterns by the Metallurgy Division. The results of these analyses are given in Table IV.

Table IV		
<u>Chemical and X-ray Analyses of Alloy Samples</u>		
Region of Sample	Chemical: Al/U	X-ray
Near aluminum surface	2.76	UAl ₃
Near uranium surface	1.94	UAl ₃ and U

It was gratifying to find such close agreement between the chemical and X-ray analyses. These results appeared to indicate that the UAl₃ alloy was formed by the diffusion of the uranium through the alloy to the aluminum surface. The composition of the black material between the alloy and the aluminum surfaces has not been determined as yet.

The phase diagram for the U-Al system indicates that, if UAl_3 exists, then UAl_2 and UAl_5 should also be present in the reaction alloy. The UAl_2 presumably would occur near the uranium surface, and the UAl_5 would occur near the aluminum surface. No trace of the latter compounds has been found in the reaction alloy as yet.

CONCLUSIONS AND RECOMMENDATIONS

1. The temperature effect on the rate at which the alloying reaction penetrates the aluminum was obtained tentatively from two direct measurements. Many more data are needed to determine the relationship accurately.
2. The alloying reaction rate seemed to be less when 99.99% pure aluminum was used than when 2S aluminum was used. More data are required for a quantitative comparison.
3. Anodization of the aluminum surface prevents the alloying reaction at 450°C and below.
4. The alloying reaction occurs wherever and whenever clean metallic surfaces of aluminum and uranium are brought into contact.
5. The extent of the alloying reaction appeared to increase with increased pressure between the aluminum and uranium surfaces. More data are needed for a quantitative relation.
6. In the alloying reaction, the aluminum and uranium form UAl_3 . The uranium diffuses to the aluminum surface through the UAl_3 layer. Many more data are required to determine accurately the mechanism of the alloying reaction. Also, the temperature effect on the reaction mechanism is unknown.
7. The role which the black substance found between the aluminum and the alloy plays in the alloying reaction should be investigated.

APPENDIX

A. Procedure Details

Polishing the Aluminum Disks

The disks were previously machined parallel and smooth. They were then polished by standard metallographic polishing procedures. The papers used were 0, 2/0, 3/0, and 4/0 grade emery polishing paper. The grit was washed off with water between papers. A carrier, a solution of paraffin in kerosene (6 gms/200 cc), was used on the polishing papers. Then the disks were polished on a polishing wheel using "Gamel" cloth, with #2 alumina abrasive and water. After an alcohol rinse on the polishing wheel, the disks were stored in absolute alcohol.

Prior to the assembling operation, the disks were polished on the "Gamel" cloth with #2 alumina and water. Then they were rinsed with alcohol on the polishing wheel and placed in the stainless steel tank filled with absolute alcohol (Figure 11).

Polishing the Uranium Disks

The disks were previously machined parallel and smooth as in the case of the aluminum. Before any polishing papers were used, the uranium disks were dipped in a 1 to 1 solution of nitric acid and distilled water. The dipping was done to penetrate the oxide film which forms practically instantaneously on exposure to the air. This characteristic of uranium made it very difficult to keep a clean, oxide-free surface. The same papers and carrier were used for the uranium disks as were used for the aluminum disks. The disks were polished on a polishing wheel using "Billiard" cloth, with a carborundum abrasive and water. After an alcohol rinse on the polishing wheel, the disks were stored in absolute alcohol.

Prior to the assembling operation, the disks were polished on the "Billiard" cloth with the carborundum abrasive and water; then they were rinsed with alcohol on the polishing wheel and placed in the stainless steel tank containing absolute alcohol (Figure 11).

Cleaning of Assembly Components

The various components of the assembly (Figure 2 and Table I) were thoroughly cleaned in order to remove any substances which might later volatilize and contaminate the aluminum and uranium surfaces. Only reagent grade solvents and absolute alcohol were used. The steps in the cleaning procedure for the assembly components are outlined below.

A. Pyrex container:

1. Washed thoroughly with soap and hot water.
2. Washed with carbon tetrachloride.

3. Rinsed with acetone.
4. Dried on sealing apparatus with He.

B. Stainless steel clamp:

1. Washed with H_2O .
2. Washed with carbon tetrachloride.
3. Rinsed with acetone.
4. Rinsed with alcohol.
5. Stored in alcohol.

C. Nickel screen basket containing uranium turnings:

1. Washed with acetone.
2. Washed with ethelene trichloride.
3. Immersed in 1:1 solution of concentrated nitric acid and distilled H_2O .
When silvered, the acid was displaced with distilled water.
4. Rinsed with acetone.
5. Rinsed with alcohol.
6. Stored in alcohol.

D. Aluminum compression cup:

1. Washed with H_2O .
2. Washed with carbon tetrachloride.
3. Rinsed with acetone.
4. Rinsed with alcohol.
5. Stored in alcohol.

E. Aluminum disk:

1. Washed with carbon tetrachloride.
2. Polished (see "Polishing the Aluminum Disks").
3. Rinsed with alcohol.
4. Stored in alcohol.

F. Uranium disk:

1. Washed with carbon tetrachloride.
2. Polished (see "Polishing the Uranium Disks").
3. Rinsed with alcohol.
4. Stored in alcohol.

G. Stainless steel shim:

1. Washed with H_2O .
2. Washed with carbon tetrachloride.
3. Rinsed with acetone.
4. Rinsed with alcohol.
5. Stored in alcohol.

Assembling the Sandwich

Most of the equipment used to assemble the aluminum and uranium sandwich is shown in Figure 10. The support on the left holds a vacuum chamber which was

used to remove the last traces of alcohol from the assembly. The stainless steel tank on the right was used to assemble the disks in the clamp and place the sandwich in the vacuum chamber beneath the surface of the alcohol medium.

After cleaning, the assembly components and the brass cover plate from the vacuum chamber were placed in the stainless steel tank containing alcohol by means of forceps (Figure 11). Rubber gloves previously cleaned and rinsed in alcohol were used to assemble the components in the tank. The disks were placed loosely in the clamp, allowing a slight space between surfaces, and the tank was raised under the vacuum chamber until the chamber was completely immersed in alcohol (Figure 12). Helium was then allowed to flow freely through the vacuum chamber. The assembly was inserted in the vacuum chamber, and the cover plate set loosely in place. The pan was then lowered while the helium flowed rapidly past the cover plate. When the alcohol stopped dripping from the chamber, the cover was bolted on and the helium turned off. The cylinder was then alternately evacuated and flushed with helium. This allowed the alcohol in the cylinder to evaporate away, leaving the assembly perfectly dry. The helium also diluted and flushed away any possible contamination from air leaks into the cylinder. This procedure was followed for approximately 1/2 hour. At this time, it was assumed that the alcohol had evaporated completely from the assembly. The helium was left on and the cover plate removed. The only opening was the clearance between the assembly clamp and the chamber wall, which was quite small (Figure 13). The positive helium pressure prevented air from entering the chamber and causing contamination of the assembly. The three nuts on the stainless steel clamp were tightened evenly by the use of a torque wrench. A torque of 20 inch-pounds was used in all cases. During the assembling operation, the glass container was cleaned and rinsed in acetone and allowed to dry on the sealing apparatus by circulating helium through it. A nickel screen basket containing uranium turnings was inserted into the tube and also allowed to dry. The pyrex container was placed in position in front of the vacuum cylinder. The helium flow remained on in both directions (from both the vacuum chamber and the pyrex container). The brass plug on the left end of the vacuum cylinder was removed and the assembly inserted into the tube with the aid of a clean rod (Figure 14). The second uranium-filled nickel basket was placed in the tube and allowed to dry. A one-hole rubber stopper covered with aluminum foil was placed in the end of the pyrex tube. The helium flowed through the tube for approximately 1/2 hour. At this time the pyrex tube was sealed at point "A," keeping a helium atmosphere in the container at all times (Figure 15). The completed assembly was then evacuated for approximately 15 hours. After this period of time, the vacuum usually ranged from 1 to 5 microns. The capillary section, point "B," in the pyrex tube was then sealed off, leaving the completed assembly.

Exceptions to the Above Procedure

In the case of Assembly #7, a uranium disk was placed on top of an aluminum disk and vice versa. A photograph of this assembly is shown in Figure 17. When Assemblies #9 and #10 were made, the nickel baskets with uranium turnings, were left out of the container, since air and helium atmospheres, respectively, were used for this test. The aluminum disks used for Assembly #12 were anodized according to the procedure used for the Brookhaven finned aluminum tubing.

Test Procedure

The completed assembly was first placed in a preheat furnace, which brought the temperature of the assembly up to the desired test temperature in a period of several hours. The assembly was transferred very rapidly to the constant temperature tube furnace (Figure 16) for the completion of the test. As mentioned previously, photographs of the assembly at the test temperature were taken periodically to observe the progress of the reaction. Figure 18 shows how an assembly looks after the alloying reaction has proceeded to an advanced stage. At the conclusion of the test, the assembly was removed from the constant temperature furnace and placed in the preheat furnace, which had been brought up to the same temperature as the assembly; then the preheat furnace was allowed to cool to room temperature. After cooling, the pyrex container was broken. Frequently, the uranium turnings were pyrophoric and had to be buried in powdered graphite. The clamp was then removed, and the surfaces were parted. The condition of the surfaces was noted carefully and photographs were taken. Apparently the combined effects of high vacuum and high temperature left the uranium surfaces in a condition not subject to the rapid tarnishing in air as is usually noted in the case of exposed surfaces of uranium metal.

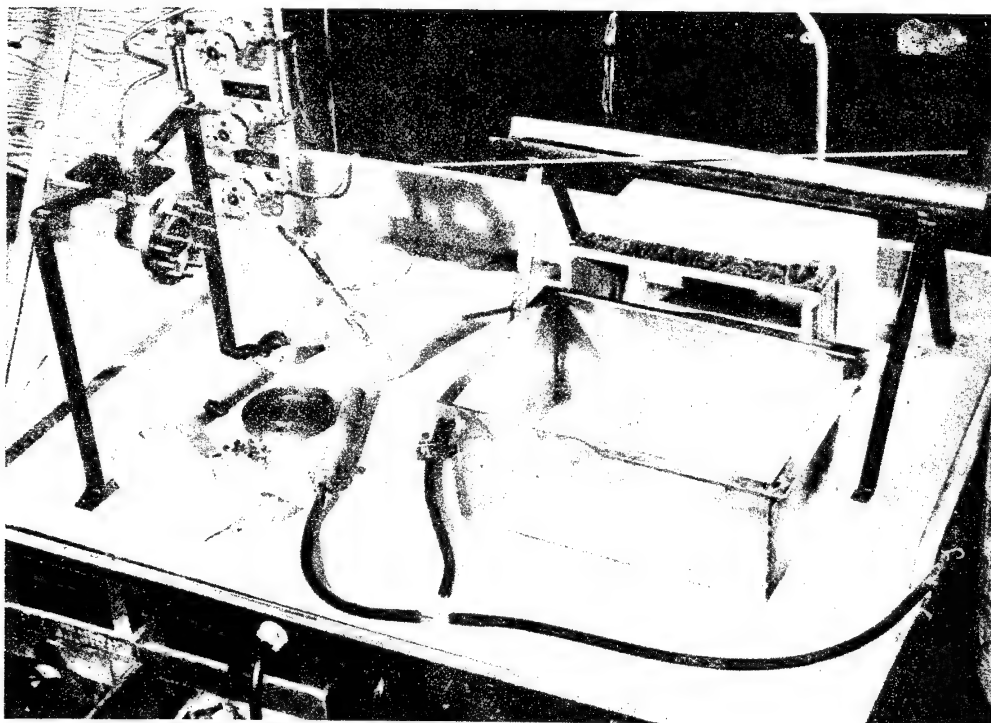


Figure 10. Equipment used to assemble the sandwich.

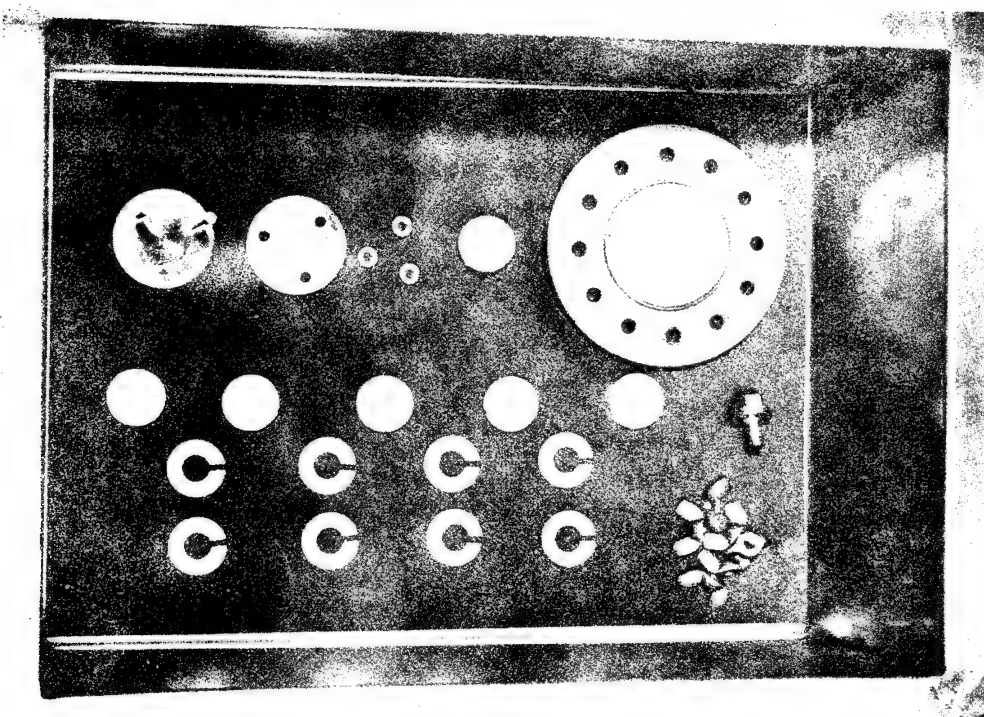


Figure 11. Assembly components in the stainless steel tank.

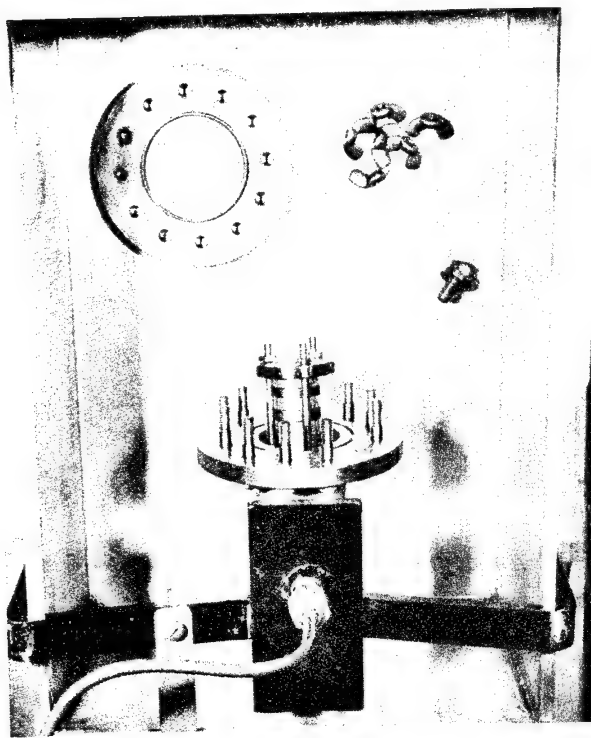


Figure 12. Assembly entering the vacuum chamber.

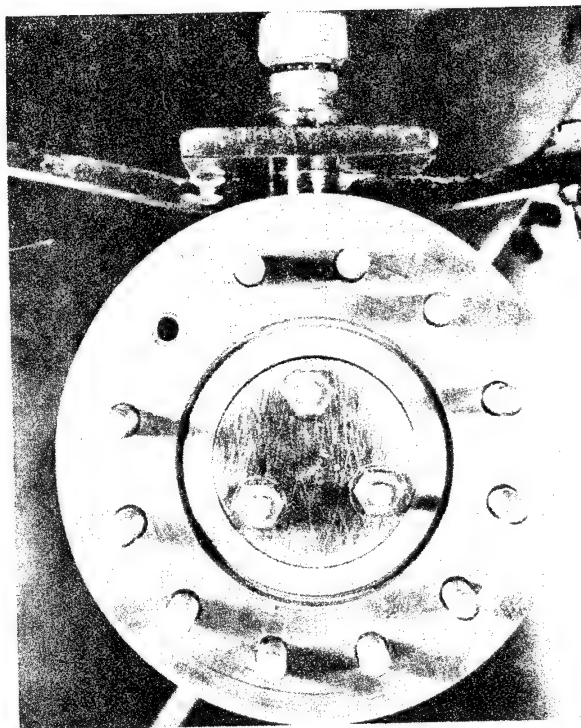


Figure 13. Assembly in the vacuum chamber.

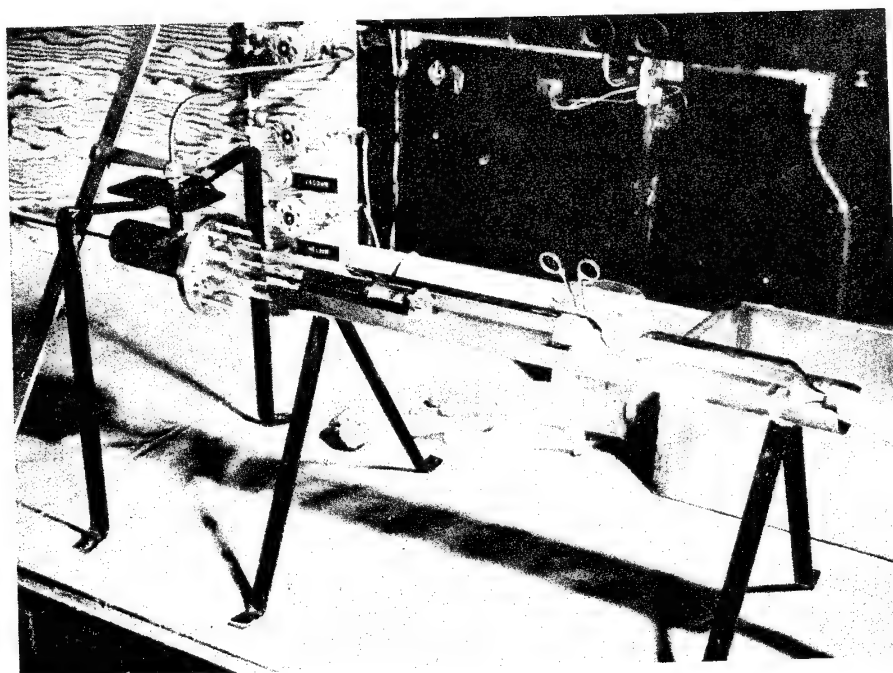


Figure 14. Assembly entering the pyrex container.

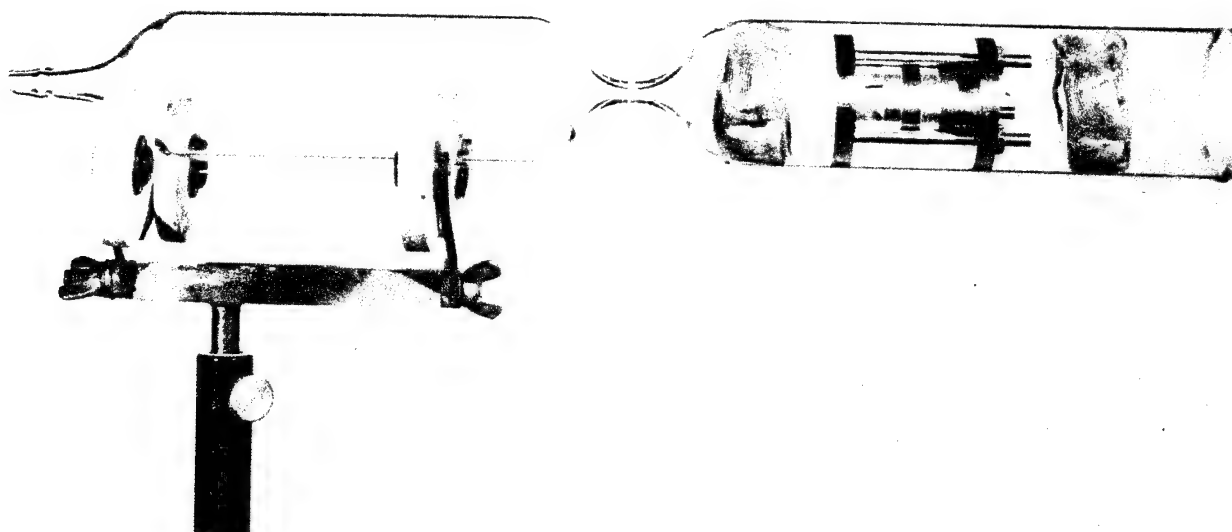


Figure 15. Assembly prepared for sealing off the container.

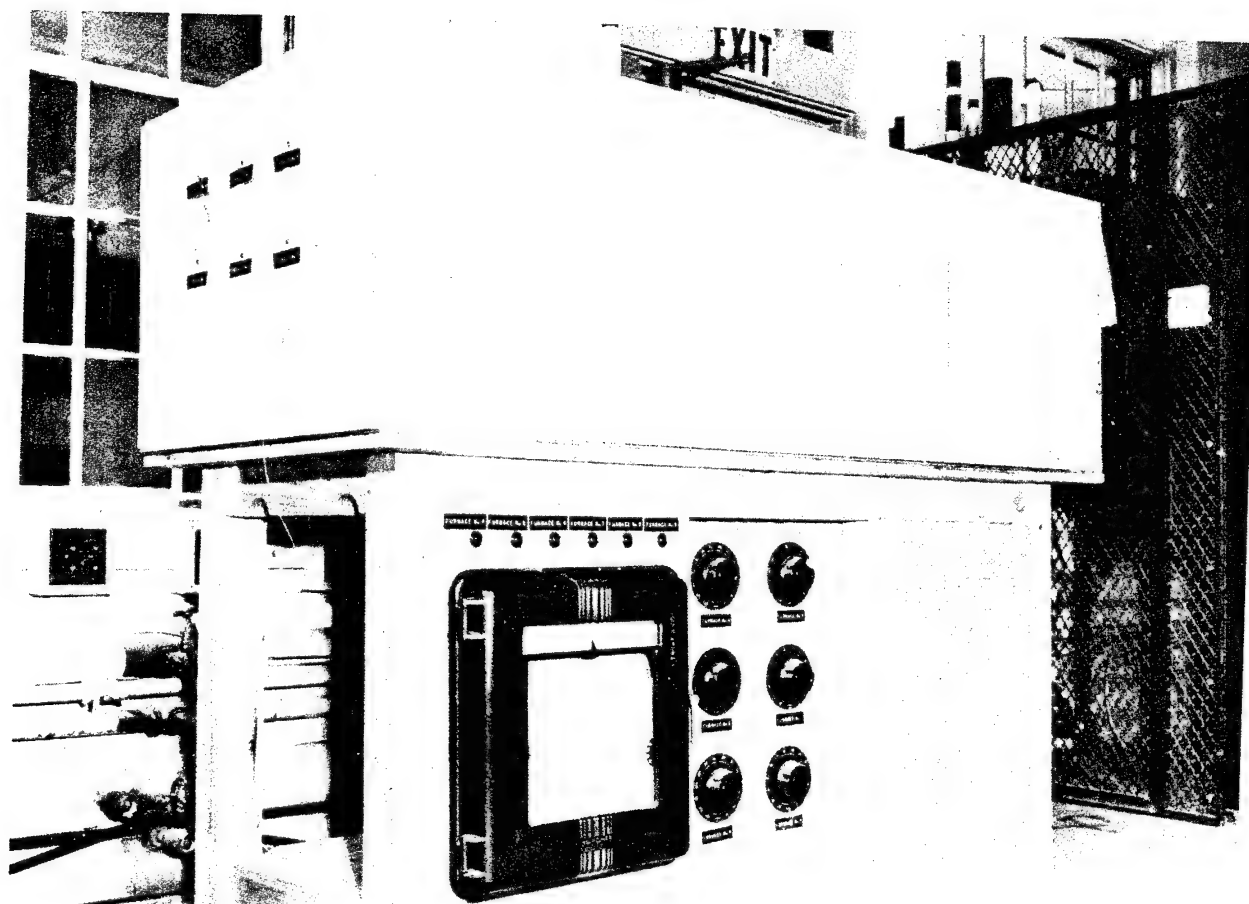


Figure 16. Constant temperature tube furnaces.

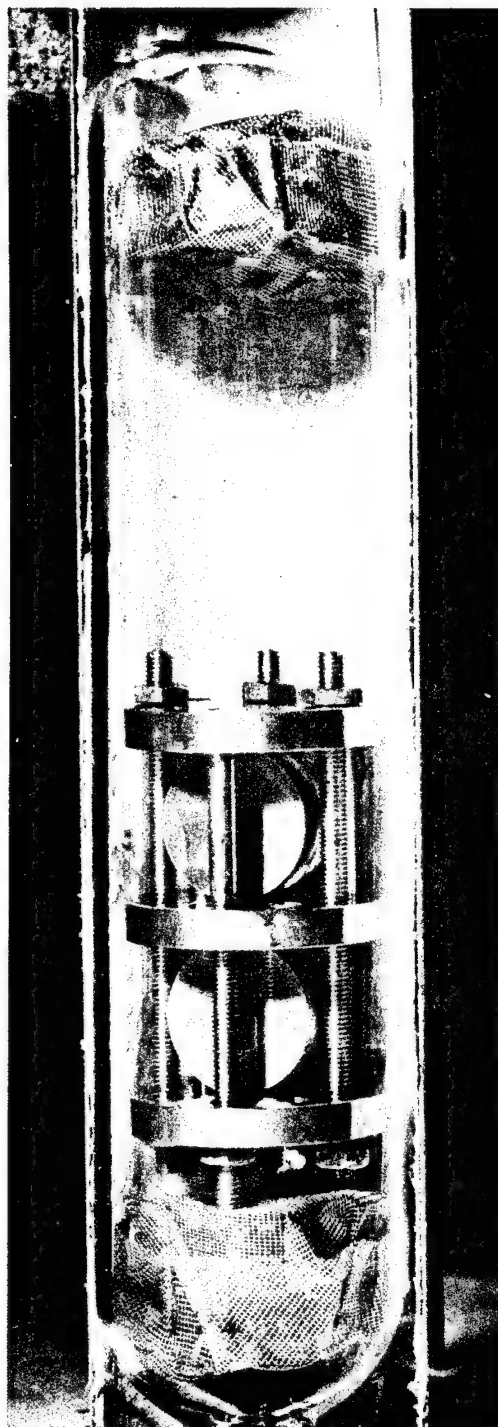


Figure 17. Assembly #7 - low interface pressure.

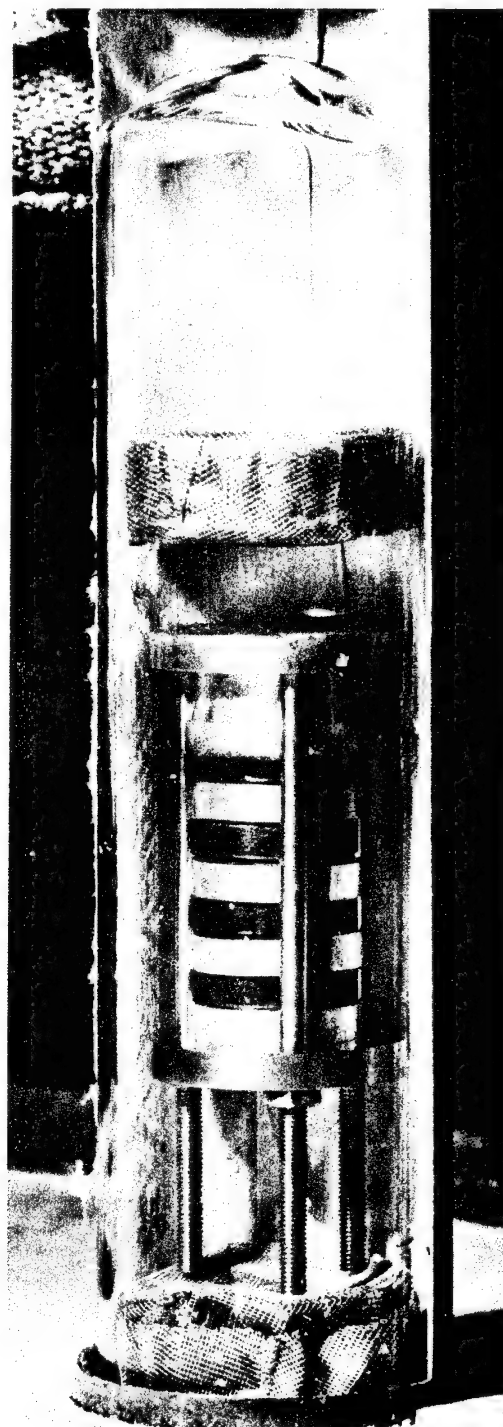


Figure 18. An assembly with advanced alloy formation (#2).

B. Photographs of Aluminum and Uranium Surfaces

While most of the surfaces of the separated assemblies were photographed, only one representative photograph from each assembly is included herein. In the following set of photographs, the uranium surface is located on the right hand side of the page (Figures 19 - 28).

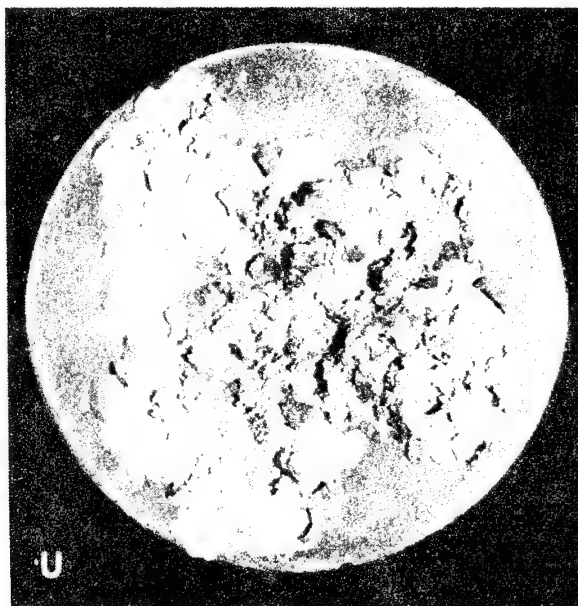
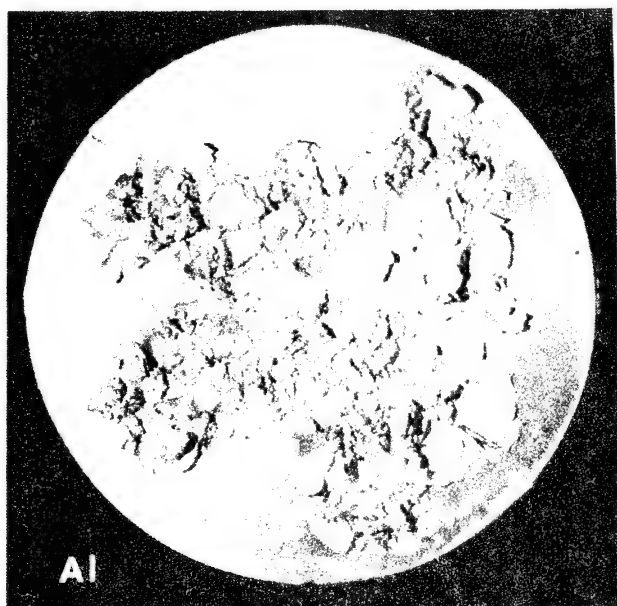


Figure 19. Assembly #1 - Interface #2.

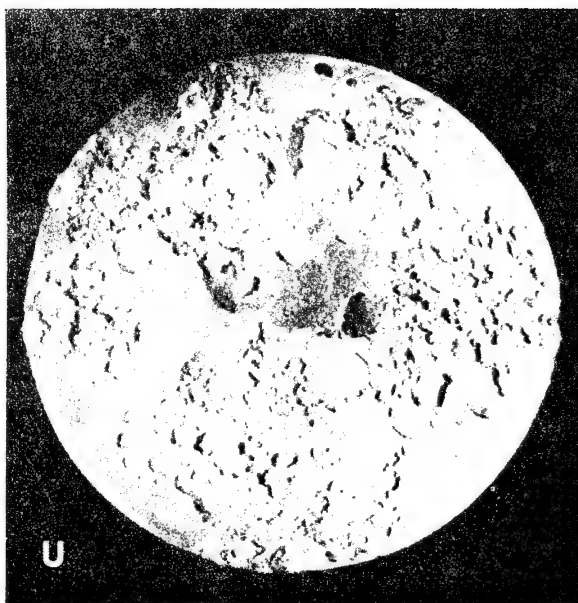
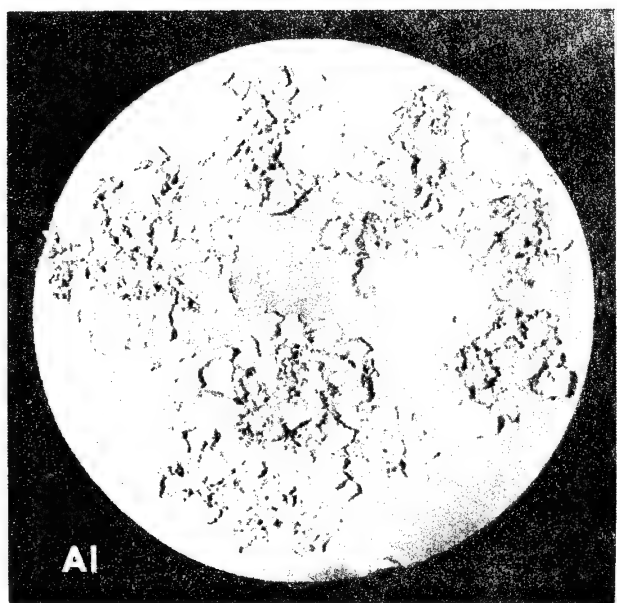


Figure 20. Assembly #2 - Interface #3.

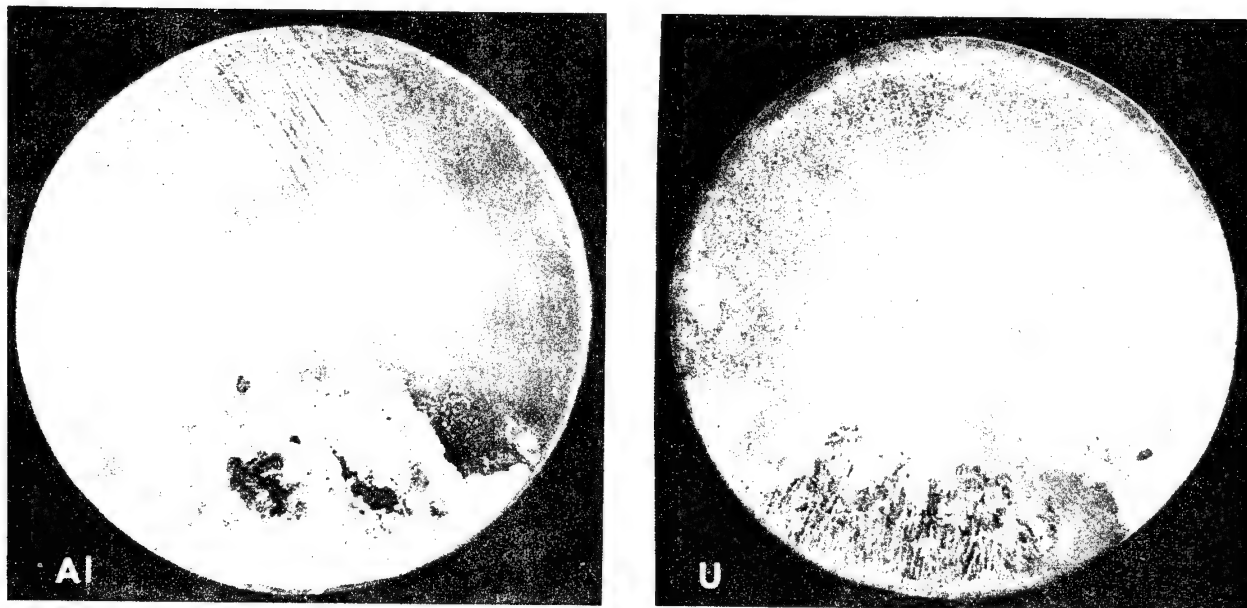


Figure 21. Assembly #3 - Interface #1.

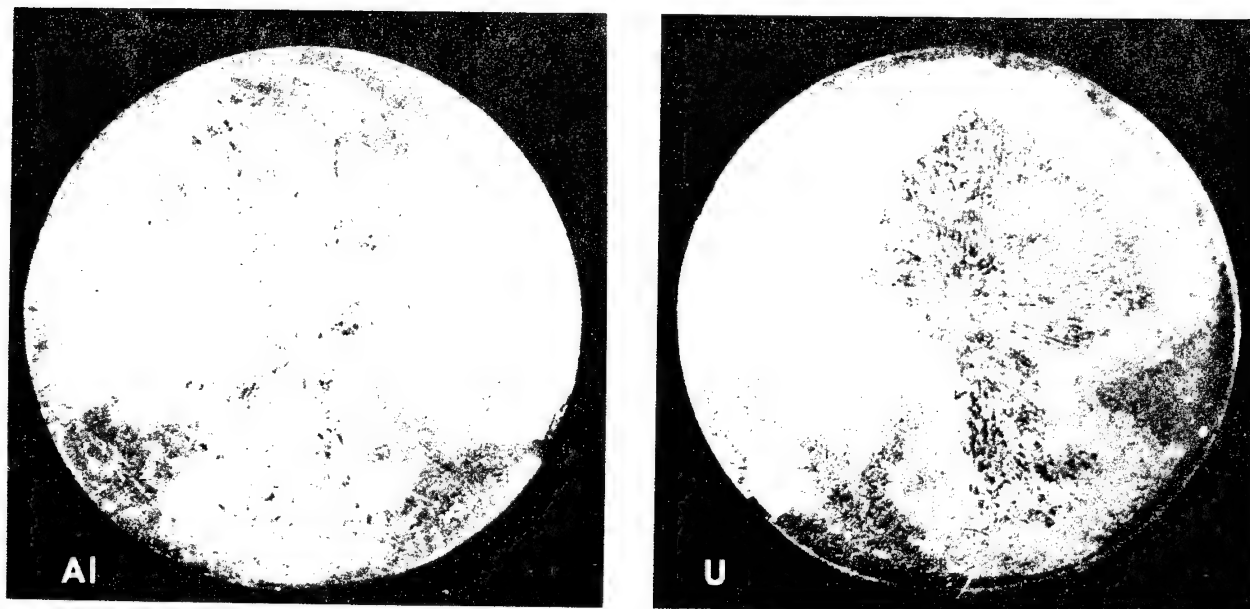


Figure 22. Assembly #4 - Interface #3.

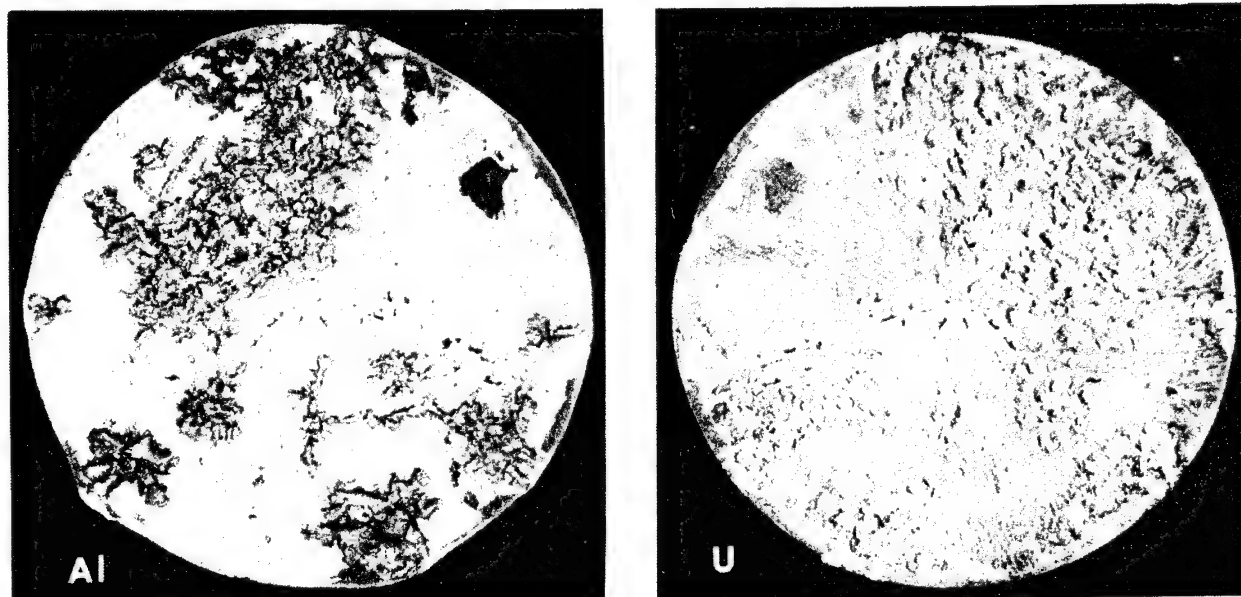


Figure 23. Assembly #5 - Interface #1.

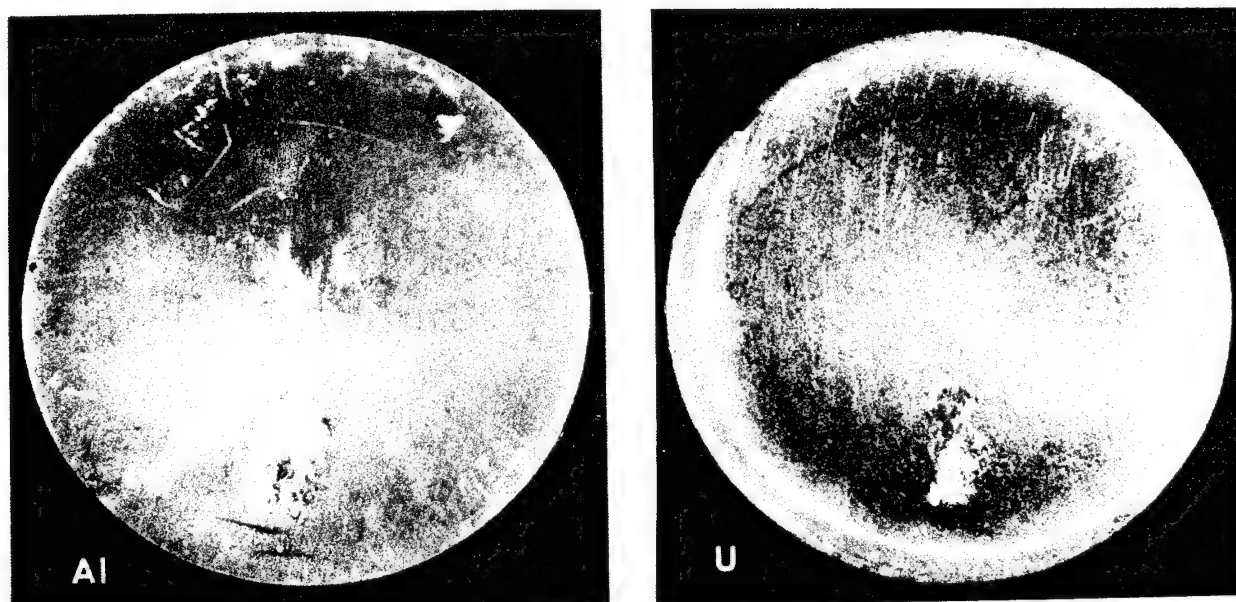


Figure 24. Assembly #7 - U on Al.

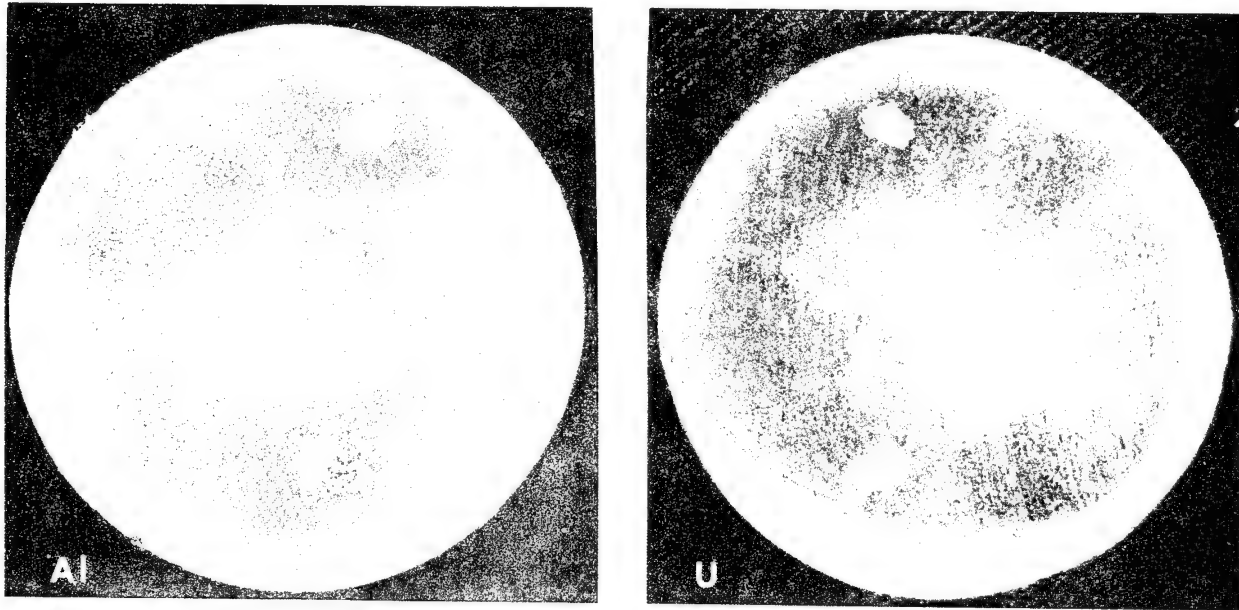


Figure 25. Assembly #7 - Al on U.

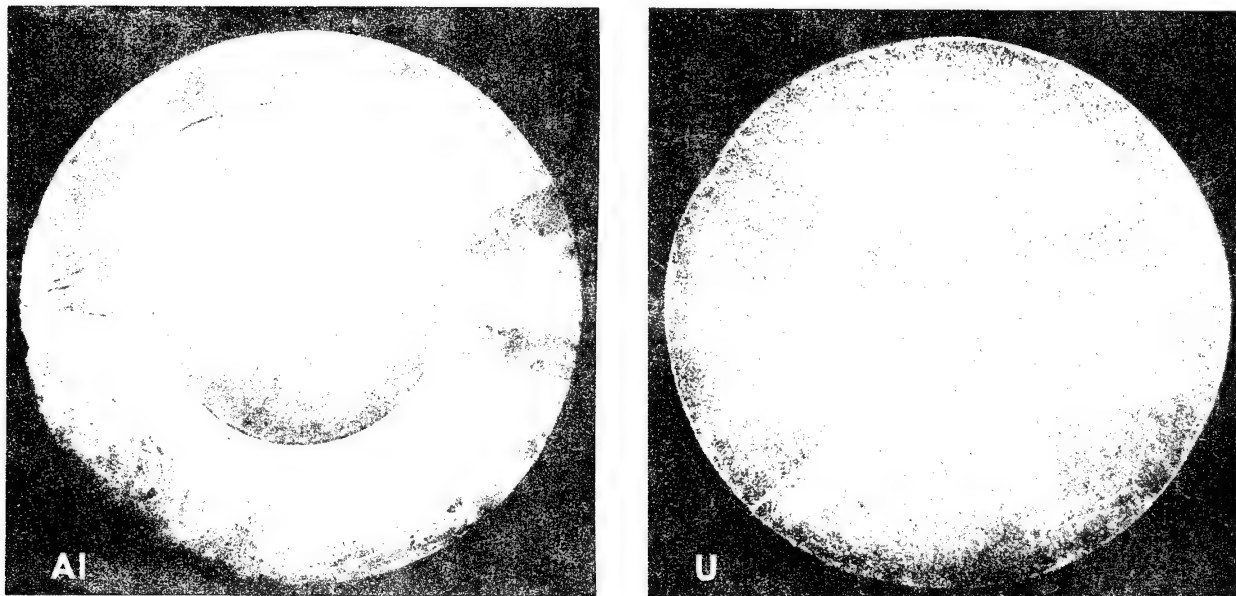


Figure 26. Assembly #8 - Interface #1.

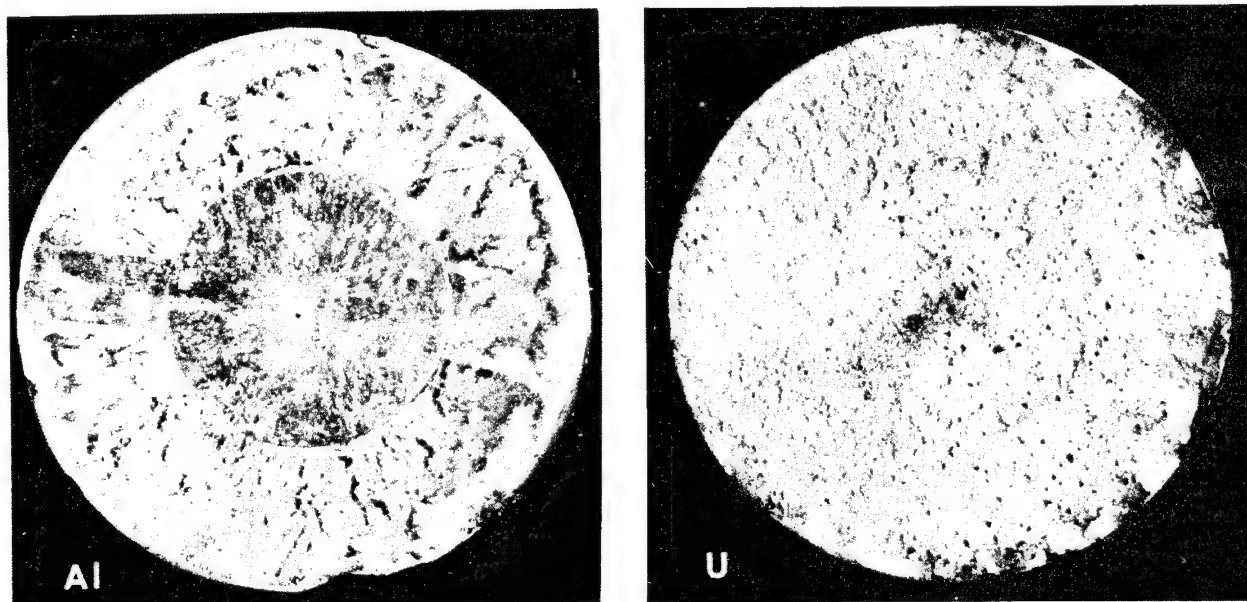


Figure 27. Assembly #9 - Interface #4.

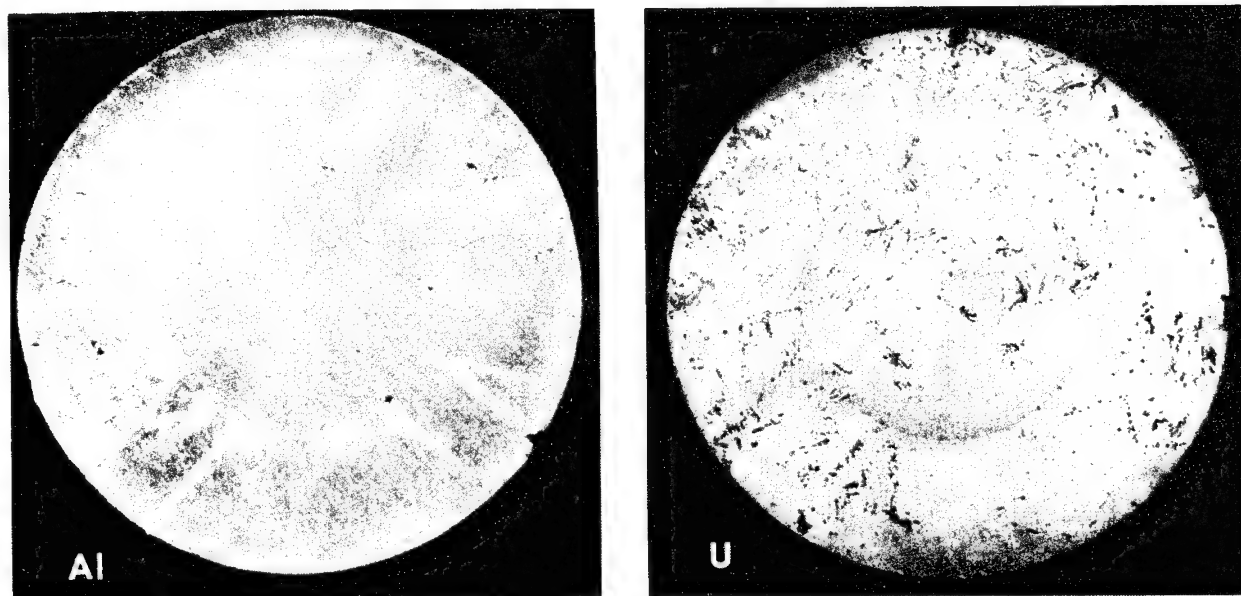


Figure 28. Assembly #10 - Interface #1.

C. Diffusion Constants

It is possible to rearrange the equations shown in Figure 9 in the form of the diffusion equation:

$$D = D_0 e^{-\frac{E}{RT}},$$

and to obtain the constants D_0 and E . These values are shown in Table V, with the units used most frequently in the literature.

Table V		
<u>Diffusion Constants Obtained from the Alloying Reaction Data at BNL and ORNL</u>		
Constant	BNL	ORNL
D_0 (cm ² /sec)	1.1	1.9×10^{-4}
E (cal/g-atom)	24,000	15,000

All of these constants fall within the range of observed values for other solid-solid systems.

D. X-ray Diffraction Patterns

The X-ray diffraction patterns obtained from the samples of alloy referred to in a previous section are shown in Figures 29 and 30. The lines in Figure 29 correspond to those obtained for UAl_3 . Figure 30 contains the UAl_3 lines plus others which correspond to those obtained for uranium metal. The alloy samples came from Assembly #1.

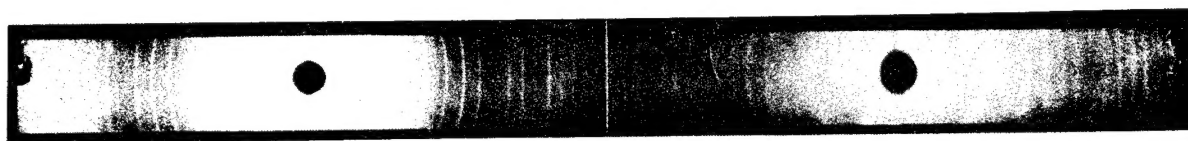


Figure 29. X-ray diffraction pattern of alloy near aluminum surface.

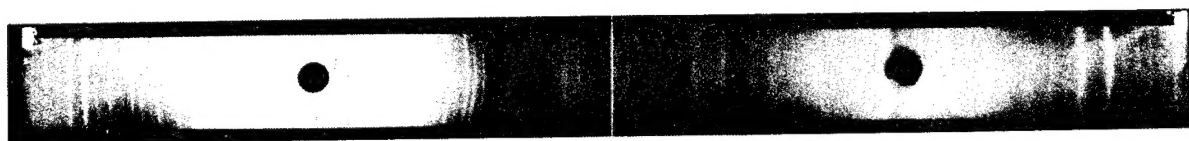


Figure 30. X-ray diffraction pattern of alloy near uranium surface.

E. Chemical Analyses

Disk Metals

The 2S aluminum probably had an analysis similar to the following, which was obtained from the manufacturer.

<u>Al</u>	<u>Fe</u>	<u>Si</u>	<u>Cu</u>	<u>Mn, Mg</u>
99.2	0.4-0.5	0.15-0.2	0.15	<0.05

The pure aluminum had an analysis as follows:

<u>Al</u>	<u>Si</u>	<u>Fe</u>	<u>Cu</u>	<u>Mg</u>	<u>Mn, Ca, Na</u>
99.99	0.0019	<0.0005	0.0004	0.0008	Not detected

The uranium used came from slugs of the type used for the Brookhaven reactor.

Reaction Alloys

Samples of uranium-aluminum alloys were analyzed as follows. Weighed portions of each alloy were dissolved in HCl and made up to volume. One aliquot of each portion was precipitated with NH_4OH , ignited, and weighed as the mixed oxides (Al_2O_3 and U_3O_8); separate experiments showed this procedure to be reliable. One aliquot of each portion was reduced with zinc amalgam and titrated with permanganate, to give the uranium content of the sample. The aluminum content was calculated from the uranium content and the weight of the mixed oxides. Due to the small amount of sample #2 which was received, the second portion of this sample was not sufficiently large for accurate gravimetric analysis; therefore, the results calculated from the total of both portions of sample #2 are given, in addition to the results from each portion

Sample No.	1a	1b	2a	2b	2 (total)
Mg sample taken	0.3394	0.4005	0.2362	0.1073	0.3435
% U	76.05	75.75	79.55	81.7	80.30
% Al	23.85	23.52	18.8	14.6	17.6
Total % U plus Al	99.90	99.27	98.4	96.3	97.9
Mol ratio, Al/U	2.77	2.74	2.09	1.58	1.94
Source	Assembly #2		Assembly #1		
Location	Near Al surface		Near U surface		

F. Acknowledgement

The author is indebted to many individuals for their valuable advice and assistance. Most of the laboratory work was performed by G.A. Schoener and M.A. Thomas. The continuous assistance given by O.F. Kammerer of the Metallurgy Division was very helpful.

G. References

1. Interim Report on Clinton Slug Ruptures. Causes and Prevention, R.O. Williams, ORNL-269
2. Recent Progress of U-Al "Sandwich" Test Reaction Studies, G.J. Snyder, MIT-GJS-1
3. Engineering Tests for the Brookhaven Reactor, W.E. Winsche et al., BNL-23
4. Report of Visit of UK and Canadian Personnel to Oak Ridge on December 9-10, 1948, J. Shilling, Jr., TID-211

H. Bibliography

The following reports refer to the aluminum-uranium alloying reaction. It is not intended to be a complete list of all such reports.

Progress Report on Metallurgy of Tuballoy to University of Chicago, H.W. Russell, BMI	CT-541
Progress Report on Metallurgy of Tuballoy to University of Chicago, H.W. Russell, BMI	CT-611
Progress Report on Metallurgy of Tuballoy to University of Chicago, H.W. Russell, BMI	CT-818
Technological Research. Metallurgy, A.B. Greninger et al.	CT-953
Recent Progress of U-Al "Sandwich" Test Reaction Studies, G.J. Snyder	MIT-GJS-1
Slug Ruptures in the Oak Ridge National Laboratory Pile, C.D. Cagle and L.B. Emlet	ORNL-170
Results of Clinton Slug Cycling Experiments, W.S. Pellini	CF-48-10-148
Technical Cooperation Program Report of Visit of United Kingdom and Canadian Personnel to Oak Ridge, December 9-10, 1948, J. Shilling, Jr.	TID-211
Interim Report on Clinton Slug Ruptures. Causes and Prevention, R.O. Williams	ORNL-269
1950 Reactor Program Analysis, J.H. Frye et al.	CF-49-8-30